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Measurement of Polybrominated Diphenyl Ethers on Hand Wipes: Estimating Exposure from Hand-to-Mouth Contact

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Estimates of exposure to the flame-retardant polybrominated diphenyl ethers (PBDEs) in dust are very poor due to limited knowledge about dust ingestion. This study was undertaken to determine if PBDEs could be measured on hand wipes, and if so, to determine the distribution of levels present on the skin surface area to provide preliminary exposure estimates from hand-to-mouth contact. Hand wipes were collected from 33 individuals residing in the United States using sterile gauze pads soaked in isopropyl alcohol. The total PBDE residue collected on the wipes ranged from 2.60 to 1982 ng, with a median value of 130 ng, or normalized to hand surface area, a concentration of 135 pg/cm^2. The fully brominated congener, BDE 209, was also detected and ranged from <DL to 270 ng with a median value of 26 ng. Congener patterns observed on the wipes were similar to patterns observed in house dust samples, consisting of congeners associated with the PentaBDE and DecaBDE mixtures, suggesting that the source of PBDEs to the hands may be dust particles. However, PBDE hand residues may also be a result of direct contact with PBDE-laden products, leading to adsorption to the skin surface oils. Repeated wipe sampling from three individuals suggests that ΣPBDE levels on the hand may be relatively consistent for some individuals but not for others. Furthermore, levels of ΣPBDEs were greater on the bottom of the hands relative to the top of the hands. Using these values we have calculated potential human exposure from hand-to-mouth contact. The median exposure estimates for children and adults are 1380 and 154 ng/day, respectively, whereas the 95th percentile exposure estimates were 6090 and 677 ng/day, respectively. These estimates are greater than dietary intake rates and suggest hand-to-mouth contact may be a key exposure route for PBDEs.

Introduction

Brominated flame retardant chemicals have been used in commercial products for more than 30 years to reduce the flammability of furniture and electronic products and increase escape time in the event of a fire. Over 175 different chemicals are used as flame retardants and a majority of these are halogenated (*1*). Polybrominated diphenyl ethers (PBDEs) are a class of brominated flame retardants that have received considerable attention due to their environmental persistence and increasing concentrations in wildlife and humans (*2–6*). In laboratory studies, this class of chemicals has been shown to elicit adverse effects on the endocrine system, particularly with regard to thyroid hormone regulation, and on neurodevelopment (*7–12*).

The primary route of PBDE exposure among the U.S. population is ambiguous at present but is most likely linked to dietary intake and/or indoor exposure (i.e., dust ingestion and/or inhalation in the home). Recent market-basket surveys (*13–15*) and models (*16*) suggest that PBDE concentrations present in food do not appear to be high enough to explain the body burdens measured in the U.S. population (*17*). Furthermore, one study compared levels of PBDEs in breast milk and house dust and found a significant correlation (*18*). Considering the high levels of PBDEs that have been measured in indoor air and house dust (*19–23*), indoor exposure may be more significant than dietary exposure. Young children are likely to receive greater exposure to chemicals found in house dust because of crawling and mouthing behavior which results in greater dust ingestion (*24*). Lead exposure and accumulation in children due to house dust exposure has highlighted this issue (*25*).

It has proven quite difficult to determine the relative amount of exposure that adults and children receive from indoor dust because most assessments rely upon an estimate of dust ingestion developed by the U.S. EPA (*26*), which assumes that an adult or child ingests anywhere from 5 to 200 mg of dust per day, an overly simplistic estimate that may be inaccurate. The U.S. EPA's estimate for adults is based on very little data and is particularly uncertain. Our study was designed to increase understanding of indoor PBDE exposure by exploring potential exposure from hand-to-mouth contact. Contact with PBDE-containing products (e.g., TVs, remote controls, cell phones) and/or house dust may lead to adsorption of these chemicals to the hand and skin oils. Adsorption of these chemicals to the hands may lead to inadvertent ingestion from transfer to food that is eaten by the hands (particularly oil foods, e.g., French fries, sandwiches, etc.), through behavior patterns that may influence the intake of PBDEs (e.g., nail biting, smoking, thumb-sucking), or through dermal absorption. To date, no studies have examined the levels of PBDEs adsorbed to dermal surfaces. The objective of this study was to determine if PBDE residues could be measured in hand wipes, and, if so, to determine the distribution of levels present in a small U.S. population. A secondary objective was to then use these results in estimating potential exposure from hand-to-mouth contact to better understand the significance of this pathway to overall human exposure.

Materials and Methods

Participants. Institutional review board approval was granted for this study prior to collection of all hand wipe samples. Participants for this study were solicited through personal communications with colleagues and neighbors of the research team in Durham, North Carolina. Thirty-three individuals from the United States participated in the study. Participants were asked to sign a consent form and fill out a short questionnaire with questions regarding age, height, weight, occupation, and place of residence. Participants were

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excluded if they had washed their hands within the last 30 min prior to sampling. We also collected repeated hand wipe samples from three participants (four samples per participant) to conduct a preliminary assessment of variability over time.

Wipe Collections. Methodology for the hand wipe collections was adapted from Gordon et al. (*27*). North brand sterile gauze pads (7.6 cm × 7.6 cm) were immersed in 3.0 mL of isopropyl alcohol (reagent grade) in a clean aluminum foil tray. All samples were investigator-collected by wiping the entire surface area of the hand, top and bottom (i.e., palm), from wrist to fingertips, including the sides of the hands and fingers. One wipe was used per hand and the two wipes collected per individual were extracted and analyzed together. Three individuals were sampled in a different manner to determine if PBDE levels were different on the top of the hand relative to the bottom (i.e., palm). For these individuals one gauze pad was used to wipe the entire surface area of the palm and bottom of the fingers for both hands, and halfway up the sides of the hands and fingers. Then a second gauze pad was used to wipe the entire surface area of the top of the hand and top of the fingers for both hands, including halfway down the sides of the hands and fingers. The two wipes were extracted and analyzed separately. Gauze pads soaked in 3.0 mL of isopropyl alcohol were used as field blanks. Most wipe samples were collected at the residence of the participant and therefore a field blank was taken at each residence. Multiple field blanks were taken at residences in which multiple hand wipe samples were collected. Samples were stored in precleaned (combusted at 450 °C for 4 h) 50 mL glass centrifuge tubes at −20 °C until analysis.

Sample Extraction. Gauze pads were spiked with two internal standards (4-fluoro-2,3′,4,6-tetrabromodiphenyl ether (Chiron, Trendheim, Norway) and ^{13}C labeled decabromodiphenyl ether (BDE 209) (Wellington Laboratories, Guelph, Ontario)) and extracted with 40 mL of dichloromethane for 20 min using sonication. The extraction process was repeated 3 times and the extracts were combined. Extracts were cleaned using 4.0 g of 6% deactivated alumina eluted with 50 mL of petroleum ether. The extract was reduced in volume using a rapid evaporation system with purified nitrogen and transferred to hexane with a final volume of 0.5 mL, and a recovery standard (4-fluoro-2,3′,4,4′,6-pentabromodiphenyl ether, (Chiron, Trondheim, Norway)) was added.

Sample Analysis. All samples were analyzed using gas chromatography–mass spectrometry operated in electron capture negative ionization mode (GC/ECNI-MS), similar to our previous analysis of dust samples (*23*). Extracts were analyzed for a suite of 32 BDE congeners ranging from tri- to decaBDE (Table 1). A 0.25 mm (i.d.) × 15 m fused silica capillary column coated with 5% phenyl methylpolysiloxane (0.25 μm film thickness) was used for the separation of BDE congeners. Pressurized temperature vaporization (PTV) injection was employed in the GC. The inlet was set to a temperature of 50 °C for 0.3 min and then a 700 °C/min ramp to 275 °C was employed to efficiently transfer the samples to the head of the GC column. The oven temperature program was held at 40 °C for 1 min followed by a temperature ramp of 18 °C/min to 250 °C, followed by a temperature ramp of 1.5 °C/min to a temperature of 260 °C, followed by a final temperature ramp of 25 °C/min to 300 °C which was held for an additional 20 min. The transfer line temperature was maintained at 300 °C and the ion source was held at 200 °C. Tri through octa-BDE congeners were quantified by monitoring bromide ions (m/z 79 and 81). All three nona-BDE congeners and BDE 209 were quantified by monitoring molecular ion fragments (m/z 486.6 and 484.6), while ^{13}C BDE 209 was monitored through m/z 494.6 and 496.6.

Quality Assurance. Recovery of F-BDE-69 and ^{13}C BDE 209 averaged 84% and 50%, respectively. Ten of the hand wipe samples and four of the field blanks were found to have very low recovery (<30%) of ^{13}CBDE 209 for unknown reasons. Therefore, BDE 209 concentrations are not reported for these ten samples. The following BDE congeners were detected in field blanks: BDE 47, BDE 99, BDE 100, and BDE 209 and averaged 1.9 ± 1.0, 1.2 ± 0.36, 0.2 ± 0.08, and 3.0 ± 3.2 ng, respectively. All samples were blank corrected by subtracting the average field blank. Limits of detection (LOD) were determined by three times the standard deviation of the field blanks. For congeners not detected in the field blanks, the LOD was set at the laboratory limit of quantification (LOQ).

TABLE 1. Summary Statistics for the 15 Primary PBDEs[a] Measured in Hand Wipe Samples (Values Represent the Total Mass (in ng) Measured on Both Hands, Top and Bottom; n = 33)

congener	detection frequency (%)	median	mean	minimum	maximum
BDE 17	88	0.27	1.16	<DL	14.1
BDE 28, 33	88	0.64	1.35	<DL	7.41
BDE 47	97	42.2	72.7	<DL	565
BDE 49	100	3.94	5.49	0.42	24.1
BDE 66	100	2.24	3.07	0.02	12.5
BDE 85, 155	97	1.33	2.90	<DL	36.8
BDE 99	100	40.5	72.2	0.9	747
BDE 100	100	7.09	13.2	0.08	142
BDE 138	73	0.17	0.60	<DL	8.59
BDE 153	97	2.6	15.8	<DL	290
BDE 154	97	2.19	4.88	<DL	59.0
BDE 183	76	0.21	0.70	<DL	8.49
BDE 209[b]	67	25.5	43.1	<DL	270
ΣBDE		129	225	2.59	1982

[a] Additional PBDEs quantified in this study which are not reported due to nondetects or low frequency of detection include: BDEs 25, 30, 71, 75, 116, 119, 156, 176, 179, 180, 181, 190, 196, 197, 201, 202, 203, 205, 206, 207, and 208. [b] BDE 209 not reported for ten of the wipe samples due to low recovery (<30%) of the ^{13}C BDE 209 surrogate standard.

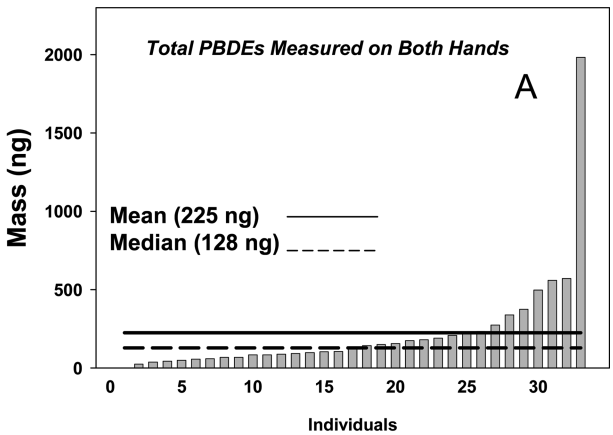
Data Analysis. Data were analyzed using SYSTAT statistical software package, version 11.0, with statistical significance defined at the $\alpha = 0.05$ level. The hand wipe data were analyzed using descriptive statistics, Pearson correlation coefficients, and t tests. A Shapiro−Wilks test was used to determine whether the log-transformed data were normally distributed. All nondetect values were treated as zero.

Results and Discussion

Study Group. Hand wipe samples were collected from 33 individuals between October 2006 and June 2007. The participating group consisted of 16 males and 17 females, including 6 children (1 girl and 5 boys) between 8 and 10 years of age.

PBDEs in Hand Wipes. PBDEs were detected on every hand wipe sample analyzed. The concentration of ΣPBDEs ranged from 2.6 to 1980 ng and the log-normalized data were normally distributed ($p < 0.05$). Table 1 presents information on the detection frequency, median, mean, and range of the dominant 15 BDE congeners measured. These values represent the ΣPBDEs measured in wipes collected from both hands top and bottom. There was no significant difference (t test on log-transformed data) in the levels of ΣPBDEs measured in wipes collected from men, women, or children, although this does not preclude differences due to the small sample size.

In general, the PBDE congener pattern was dominated by congeners associated with the PentaBDE and DecaBDE commercial mixtures (*28*). The fully brominated congener,



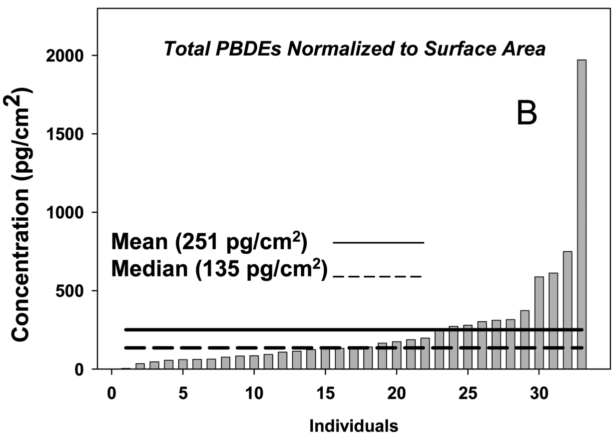


FIGURE 1. Distribution of total PBDEs (A) and surface-area-normalized concentrations of PBDEs (B) measured in hand wipe samples.

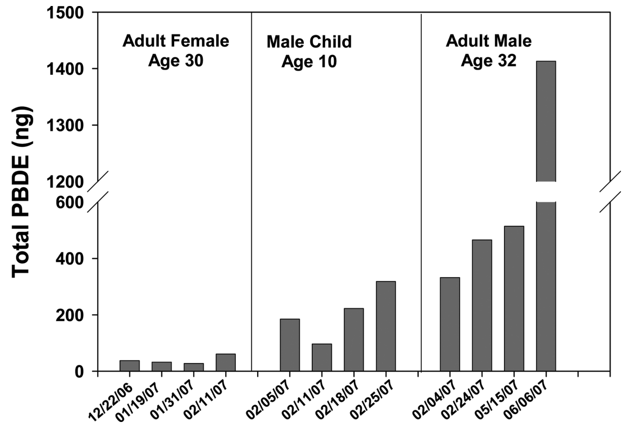


FIGURE 2. PBDE levels measured in repeated wipe collections from three individuals.

BDE 209, was detected in 67% of the hand wipe samples. The mean and maximum mass of BDE 209 measured were 43 and 270 ng, respectively. On average BDE 209 contributed 20% of the ΣPBDE mass; however, in one wipe sample BDE 209 represented 94% of the ΣPBDE mass. The PBDE congener patterns observed are very similar to patterns observed in indoor dust samples (*20, 21*) and may suggest the source of the PBDEs is from indoor dust. However, it is also possible that PBDEs may directly partition into the surface skin oils from direct contact with PBDE-laden products found in the home, such as remote controls or furniture.

Surface Area Normalization. The wipe measurements provided an estimate of total PBDE loadings on the hands; however, it was unclear whether or not the mass collected was dependent upon the surface area of the hands. We therefore normalized PBDE loadings to hand surface area to control for variability in hand surface area across participants. Surface area of the hands was estimated using a calculation from the U.S. EPA Exposure Handbook (*29*), which is based on height and weight measurements. The calculation is:

$$\mathrm{SA} = a \times \mathrm{weight}^{b} \times \mathrm{height}^{c} \times 10000 \quad (1)$$

where SA represents surface area (in cm^2), and weight and height are reported in units of kg and cm, respectively. The variables *a*, *b*, and *c* represent gender-specific constants from U.S. EPA's regressions and the value of 10,000 represents a conversion from meters to centimeters.

Surface-area-normalized PBDE concentrations ranged from 3.3 to 1970 pg/cm^2 with a median value of 135 pg/cm (*2*). Figure 1 presents the distribution of loadings and surface-area-normalized concentrations of PBDEs measured from the hand wipe samples. Normalizing to surface area did not reduce any of the variability in the measurements. The loadings and surface-area-normalized concentrations were highly correlated ($r = 0.99$; $p < 0.001$). Thus the variability in PBDE levels is not attributed to differences in hand surface area and is likely a result of behavior or exposure differences.

Repeated Wipe Measurements. As a very limited investigation of temporal variability in PBDE loadings, we asked 3 individuals to consent to repeated hand wipe collections (4 samples per individual collected over a 2-month period). The three individuals who participated were a 30-year-old female, a 10-year-old boy, and a 32-year-old male residing in different households. PBDE masses measured from repeated wipe collections are presented in Figure 2. Values from the 30-year-old female were relatively consistent and ranged from approximately 28 to 60 ng ΣPBDE. The 10-year-old boy had more variable measurements ranging from 97 to 318 ng ΣPBDEs. Lastly, the 32-year-old male had the most variable measurements which ranged from 330 to 1400 ng ΣPBDEs. Unfortunately, due to scheduling conflicts, the last two wipe samples from the 32-year-old male were taken 3 and 4 months after the first sample collection. This may be a factor in the variability observed. These results suggest that some individuals may have more consistent PBDE hand wipe loadings than others, which may depend on behavior and variables such as time of day/week and/or time since last hand-washing event. However, this investigation was very limited with 3 individuals and more studies are needed to fully examine temporal variability.

PBDEs on the Top vs the Bottom of the Hands. We also conducted a preliminary investigation to examine whether PBDE loadings are higher on the palm (bottom of the hands) relative to the top of the hands. This may be hypothesized to occur if direct contact with dust or PBDE-laden products results in partitioning to the skin. To accomplish this task, we collected separate wipe samples from the top and palm of the hand from the same individuals participating in our repeated wipe measurements. To collect the wipe samples, one wipe was used to collect the top portion of the hand from wrist to fingertips of both hands. A second wipe was collected from the palm and bottom portion of the fingers of both hands. Each wipe was analyzed separately. The proportions of ΣPBDEs measured on the top relative to the palm of the hand are presented in Figure 3.

In general, the bottom of the hand contained a higher fraction of the ΣPBDE loadings. However, this study is limited in that wipe samples collected separately from top and bottom of the hands were only taken from 3 individuals at various times. A paired *t* test was used on the log-transformed data to test the hypothesis that there were no differences in the proportion of ΣPBDEs present on the palm (mean = 0.63) relative to the top of the hands (mean = 0.37). The *p*-value, 0.066, is suggestive of a difference. Thus direct

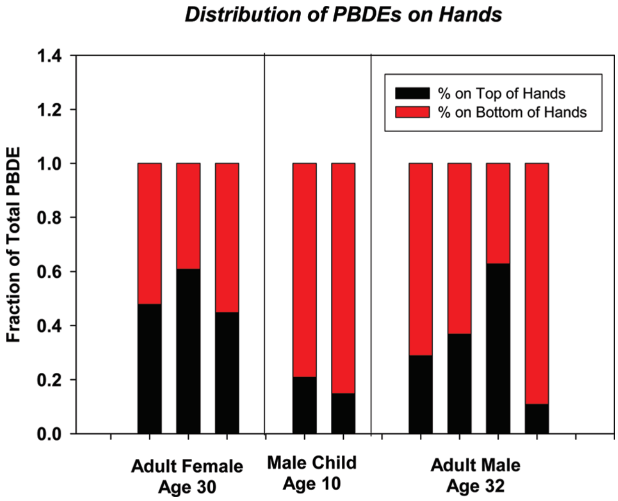


FIGURE 3. Fraction of Σ PBDEs measured on top vs bottom of hands.

contact with either dust and/or products containing PBDEs may be contributing to the levels measured on the hands, and not, for example partitioning or deposition from the air.

Exposure via Hand-to-Mouth Contact. Because PBDEs are present on the hand surface area, hand-to-mouth contact has the potential to increase human exposure through inadvertent ingestion. Any behavior that increases the frequency of hand-to-mouth contact, such as smoking, nail biting, or eating finger foods (i.e., French fries, sandwiches, etc.), may lead to increased exposure to these chemicals. Dermal absorption may be important as well. Previous laboratory studies with mice demonstrated that 62% of a BDE 47 dose was absorbed through the skin when applied to the skin in acetone (*30*). However, it is unclear how well PBDEs in dust would be absorbed through human skin, and thus will not be addressed here.

Previous dust exposure estimates for both children and adults assume that an individual ingests a known mass of dust per day through a variety of pathways that include consumption of dust-contaminated food, respiration of dust particles, and hand-to-mouth contact (*31, 32*). Previous research studies have attempted to estimate adult ingestion of PBDEs from dust using dust intake rates ranging from 0.56 to 50 mg/day (*16, 21, 33*). With median values of about 4250 ng of total PBDE/g dust (*21*), these uncertain dust ingestion rates would equate to 2−212 ng ΣPBDE ingested per day, a wide range.

Using the hand wipe data we can improve our estimates of exposure and specifically address exposure from hand-to-mouth contact. Children's exposure to a variety of chemicals and pesticides from hand-to-mouth contact activities has been a common area of study and numerous investigations have attempted to measure children's hand-to-mouth frequencies (*35*). For this study we have modified an exposure calculation from Hubal et al. (*24*). Here we will define exposure from hand-to-mouth contact by the following equation:

$$E_{derm} = M_{surf} \times TE \times SAC \times EF \qquad (2)$$

where E_{derm} is exposure (ng/day), M_{surf} is total PBDE mass present on the hands (ng), TE is transfer efficiency (%) or fraction of PBDE mass transferred at each contact, SAC is the proportion of the hand area contacted in each event (%), and EF is frequency of contact over a 24-h period (day^{-1}). Using this equation we can estimate exposure from hand-to-mouth contact using the hand wipe measurements as M_{surf} and include estimates from previous studies for other factors.

Quite a few papers have examined children's hand-to-mouth contact frequency (EF) and the numbers are variable depending upon the method used (*34–36*). For this study we used the values reported by Tulve et al. (*35*), which found that children ages 0−4 yr had an average frequency of 16–18 hand-to-mouth contact events per hour. No information on adult hand-to-mouth contact frequencies could be found, but here we will use data on contact frequency for older children (ages 6–13 yr) in the 25th percentile identified by Zartarian et al. (*37*), 2 contacts per hour. However, this may be a low value for adults prone to smoking and nail biting or other behaviors increasing hand-to-mouth frequencies. Estimates for the transfer efficiency, TE, a measure of the tendency for PBDEs to transfer from the hand to the mouth are unknown. Given the range in physical chemical properties (i.e., log K_{ow} and vapor pressure) (*38–40*) among PBDE homologue groups, it is to be expected that transfer efficiencies will also vary with bromination. Ingestion may also be influenced by the solubilization of PBDEs in saliva. However, consumption of dietary food items, particularly finger foods high in fat content (e.g., French fries, pizza, chicken nuggets, etc.) would probably accelerate the transfer of PBDEs from oral skin surfaces to food items resulting in ingestion. To estimate transfer efficiency (TE) of PBDEs from hands to mouth we replied on a report by the U.S. EPA which describes a model (SHEDS) for examining children's exposure to CCA (chromated copper arsenate) treated wood found in playsets (*37*). Their model used a value of 70% for transfer efficiency of CCA during hand-to-mouth contact. For the purposes of our estimate, we will use a lower value of 50% to account for physicochemical differences between CCA and PBDEs. We used an SAC value of 10% for both children and adults, also derived from the SHEDS model. Here we will not consider hand washing since we cannot account for the removal efficiency and subsequent kinetics of readsorption of PBDEs to the hand surfaces.

Using these variables, we have estimated the potential exposure from hand-to-mouth contact for children and adults using the median and 95th percentile values measured on the hand wipes. Table 2 presents the variables used in this model and the exposure estimates relative to reported exposures from dust and diet. Median daily exposure to PBDEs from hand-to-mouth contact was calculated to be approximmately 1400 and 154 ng/day for children and adults, respectively. Using the 95th percentile values for PBDE residues in hand wipes, the estimate is 6090 and 677 ng/day for children and adults, respectively. The exposure estimates calculated here are higher than reported exposure estimates based on diet (*14, 41*) and in the case of children, comparable to what has been reported for breast milk ingestion by infants (*42*). Relative to previous dust ingestion estimates median exposure estimates from hand contact is in some cases comparable (*16*), or higher (*21, 33*). However, the 95th percentile estimates using dust ingestion rates are higher than rates calculated for hand-to-mouth contact, reflecting some high PBDE measurements made in house dust. Thus models estimating total exposure to PBDEs using median dust ingestion rates may be underestimating the true exposure from hand-to-mouth contact, for both adults and children.

Previous studies conducted by McDonald (*17*) and Lorber (*16*) have attempted to calculate the PBDE intake rates necessary to sustain serum PBDE levels measured in the U.S. population. Their calculations suggest average individuals must be exposed to more than 500 ng of ΣPBDEs/day to reflect current serum levels. Thus the combined exposure from diet and hand-to-mouth contact reported here (∼200 ng/day) accounts for almost half of this value. However, there are many uncertainties in our model, particularly the frequency of hand-to-mouth contact for adults and the

TABLE 2. PBDE Exposure Estimates from Hand-to-Mouth Contact and Variables Used in Calculating the Exposure

variable	child age 1–4	adult	reference
contacts per hour	18	2	*34, 35*
transfer efficiency (%)	50	50	assumed
fraction of hand contacted	0.1	0.1	*37*
hours per day exposed	12	12	assumed

exposure estimates (ng/day)

population	exposure route	50th percentile	95th percentile	reference
child (>48 months)	hand to mouth	1380	6090	this study
child	dust ingestion[a]	590	1800	*21*
child	dust ingestion[b]	230	19,000	*33*
child	dust ingestion[a]	737	18,700	*23*
child	diet	28.6	263	*14*
adult	hand to mouth	154	677	this study
adult	dust ingestion[c]	59	180	*21*
adult	dust ingestion[d]	95	5000	*33*
adult	dust ingestion[e]	354	NA[f]	*16*
adult	dust ingestion[c]	74	1870	*23*
adult	diet	39.2	296	*14*
infant	breast milk[g]	1800	9000	*42*

[a] Assuming a child ingests 100 mg dust per day. [b] Assuming a child ingests 50 mg dust per day. [c] Assuming an adult ingestion of 10 mg dust per day. [d] Assuming an adult dust ingestion rate of 20 mg dust/day. [e] Assuming an adult dust ingestion rate of 50 mg dust/day. [f] NA indicates not available. [g] Assuming an infant ingests 800 mL of breast milk per day.

transfer efficiency of PBDEs with each contact, which could be responsible for these differences.

It is also important to note that we are assuming children will have the same PBDE residue concentrations on their hands relative to adults. Six children ages 8−10 were examined in this study; however, one might expect young children (e.g., less than 4 years of age) to have higher PBDE residue levels given their predilection for crawling and playing on the floor. These results warrant further studies to determine levels of PBDEs on children's hands and the transfer kinetics and bioavailabilty of these flame retardants from hand-to-mouth contact.

This study has demonstrated that PBDEs are adsorbing to the hand surface area. Furthermore, improvements on PBDE exposure estimates can be made by modeling hand-to-mouth contact instead of relying upon simplistic estimates of dust ingestion rates. Regardless of how these chemicals are adsorbing to the hand surface areas, either from dust particles or direct partitioning from contact with products, the estimated transfer of PBDEs from hand-to-mouth contact is not trivial, and is likely a significant exposure route. In addition, this study also highlights the importance of determining children's body burdens and understanding potential health effects, particularly since these compounds may elicit greater adverse effects during sensitive developmental stages.

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